

REMARKS

*Support for the Claim Amendments*

The added phrase in the amended claim 1 is based on the description in the "6) Modification ratio" on page 42.

The additional features in the new claims 27 and 28 correspond to those in claims 2 and 3, respectively.

*Invention of Claims 1-15*

Since the analysis method has been limited to that specifically disclosed in the Examples, Applicants consider that the issue of new matter should be withdrawn.

The Examiner indicates that the proposed amendment (the incorporation of the limitation of claim 9 into claim 1) in Applicants' response dated February 19, 2004 does not place the application in condition for allowance, since the Examiner has rejected claim 9 by the combination of the prior art of record (i.e., JP '959 and JP '161).

In this regard, it has been explained that the feature of the present invention particularly resides in that component (A-1) has a modified component as high as exceeding 60 wt.%, which had not been realized in the prior art technique. To describe this feature more clearly, Applicants have added to claim 1 the phrase "adsorption amount analysis with GPC using a silica gel as a filler", which is a new measurement method.

The important point is that under a circumstance where a sufficient amount of a modifying agent is present, the modification ratio is not determined by the amount of the modifying agent added, but determined by the ratio of the terminals that have been made to active metal terminals among the terminals of the polymer, i.e., the activity of the polymer. In the present invention, a new measure is applied to

the polymerization stage and a measurement method of modification ratio that was newly developed is employed. Thereby, a rubber composition in which component

(A-1) has a high modification ratio as high as exceeding 60 wt.% was accomplished. Neither of the cited references discloses or suggests a rubber composition in which component (A-1) has such a high modification ratio. Owing to the use of the polymer having a high modification ratio, the present invention provides a high performance composition having excellent processability even with a polymer having a narrow molecular weight distribution, whereas the conventional composition using silica as a reinforcing agent had suffered from insufficient processability, in the case of using a polymer having a narrow molecular weight distribution ( $M_w/M_n$  of less than 2.2).

The Examiner alleges that Example H of JP '959 (Table 1: what is called as "Example H" by the Examiner is correctly "SBR H") teaches a vinyl content of approximately 87%, and that the addition of approximately 0.87 equivalents of glycidyl compound would have been obvious (Paragraph 1 in the Office Action dated October 21, 2003). Further, in the Advisory Action dated March 4, 2004, the Examiner alleges that the 87% vinyl content is a percentage of vinyl in the rubber of Example H based on the total molecular weight and that this was calculated for 39.2 % content of vinyl where the  $M_w$  of 45 is normalized to 100%, since the total of vinyl and styrene has to add up to 100%. However, from a technical point of view, this Examiner's calculation and allegation, particularly the normalization of the content value based on the molecular weight ( $39.2 \times (100/45)$ ), appears to be erroneous. Applicants believe that the Examiner's allegation is technically incorrect and that the cited reference does not disclose or suggest a vinyl content of 87%. This is explained in detail below.

The relevant portion of JP '959 discloses SBR H having a styrene content (S) of 14.8 mol% and a vinyl bond amount (V) in the butadiene unit of 39.2 mol%. This 39.2 mol% indicates a vinyl bond percentage in terms of mol in the butadiene unit of SBR H, and hence remains unchanged irrespective of the molecular weight of the SBR. Thus, the normalization as made by the Examiner

is technically incorrect. Furthermore, the Examiner's calculation is based on the asserted Mw value of 45. However, the Mw value shown in Table 1 is Mw ( $10^4$ ), which appears to be intended as Mw ( $10^4$ ). Thus, the actual molecular weight is not 45. Also in this respect, the Examiner's calculation has no basis.

One of the present inventors conducted comparative experimentation using the same polymer as SBR H disclosed in JP '959, i.e., a polymer (Sample T1) obtained by adding a predetermined amount of tin tetrachloride to a polymer obtained through common batch polymerization to carry out coupling. (Details of the comparative experimentation are shown in the attached Rule 132 Declaration.)

Sample T1:

Bonding styrene amount: 25 wt.% (14.8 mol%)

Vinyl bond amount in butadiene unit: 39 mol%

Mw: 450,000

Mw/Mn: 1.4

Coupling ratio: 48 wt.%

Modification ratio determined by silica GPC adsorption amount: 0 wt.%

In addition, another experiment was carried out using a polymer (Sample T2) obtained by, after being polymerized in the same manner as for Sample T1, adding TGAMH (a modifying agent used in the present invention) in an amount that gives excess epoxy groups with respect to the active ends of the polymer and that gives the equivalent epoxy group with respect to the lithium amount of the fed initiator, to react with each other.

**Sample T2:**

Bonding styrene amount: 25 wt.% (14.8 mol%)

Vinyl bond amount in butadiene unit: 39 mol%

Mw: 455,000

Mw/Mn: 1.4

Coupling ratio: 61 wt.%

Modification ratio determined by silica GPC adsorption amount: 55 wt.%

As shown above, the modification ratio of the polymer was not raised to the extent presently claimed, even when the modifying agent was added in an excess amount. Incidentally, Applicants presume that the difference in the coupling ratio and the modification ratio is attributable to the occurrence of a two-molecular reaction among the polymer molecules.

Using the above-mentioned samples, the evaluations described in the specification were made. For the evaluations, compositions were prepared in accordance with the blending formulation S-1. The results obtained are as follows:

	<b>Modification ratio</b>	<b>Modifier amount (equivalent/active lithium)</b>	<b>Fuel-cost-saving property (tan<math>\delta</math>, 50°C)</b>	<b>Wet skid resistance (tan<math>\delta</math>, 0°C)</b>
Sample T1	0	0	0.200	0.505
Sample T2	55	1.54	0.185	0.538
Sample K (Ex. 3-4)	78	0.95	0.138	0.553

As can be seen from the above results, the composition using Sample T1 which was prepared by coupling using tin tetrachloride and which is equivalent to SBR H of JP '959, had extremely deteriorated performances.

In addition, Sample T2, which was prepared from Sample T1 with an excess amount of a modifier used in the present invention, had a lower modification ratio than claimed. The composition using Sample T2 showed considerably deteriorated performances as compared to Example 3-4 using Sample K according to the present invention.

For the above reasons, Applicants believe that the present invention is not obvious from JP '959 in view of JP '161.

In view of the experimentation results, applicants assert the following conclusions.

(I) SBR H of JP '959 does not have a modification ratio as claimed in the present application.

(II) Even when SBR H is subjected to a modification reaction using an excess amount of a modifier, the resulting polymer still does not fulfill the claimed modification ratio requirement. This shows or supports that a high modification ratio cannot be necessarily obtained merely by using a large amount of modifier.

(III) The rubber composition using a modified polymer as claimed is unexpectedly superior as compared to compositions using SBR H of JP '959 or using a modified SBR H with an excess amount of modifier.


If the Examiner has any questions regarding this Amendment, she is invited to contact Applicants' representative, Sanford B. Astor, at telephone number (310) 209-4400.

If necessary, the Commissioner is hereby authorized in this, concurrent, and future replies, to charge payment or credit any overpayment to Deposit Account No. 02-2448 for any additional fees required under 37 C.F.R. § 1.16 or under 37 C.F.R. § 1.17; particularly, extension of time fees.

Respectfully submitted,

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By:

  
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Attachment: Declaration of Haruo Yamada (Exhibit A)



Exhibit A

PATENT APPLICATION

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of

Akira SAITO et al.

Group Art Unit: 1714

Application No. 09/856,845

Examiner:  
Katarzyna Wyrozebski Lee

Filed: May 25, 2001

For: RUBBER COMPOSITIONS

DECLARATION UNDER 37 CFR §1.132

Honorable Commissioner of Patents and Trademarks  
Washington, D.C. 20231

Sir:

I, Haruo Yamade, do declare and state that:

I graduated from Niigata prefectural Nakajo high school, Department of Industrial Chemistry, in 1967.

In 1967, I was employed by Asahi Kasei and assigned to the synthetic rubber factory of said company.

Since that time I have been principally engaged in the work relating to synthetic rubbers such as polymerization, analysis and evaluation of solution polymerization SBR using an organic lithium initiator: specifically, engaged principally in analysis work in the research section for about five years, in polymer production work in the production section for about seven

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Declaration Under 37 CFR 1.132

years, and in polymer development and evaluation work in the technology and development department for about 25 years.

I am a co-inventor of the invention described and claimed in the above-identified application and am familiar with the Office Action dated October 21, 2003 issued therein.

The following comparative experimentation was conducted by me or under my supervision to demonstrate that SBR\_H disclosed in JP 7-330959 (JP '959) does not have a modification ratio as claimed and that the rubber composition of the present invention is unexpectedly superior to a composition using SBR H.

#### EXPERIMENTATION

##### 1. Preparation of Polymer

A thermostatic autoclave having an internal volume of 10 liter and equipped with a stirrer and a jacket was used as a reactor. To the reactor, 645 g of butadiene which was prepared by removing water content by distillation from a common butadiene containing impurities, 250 g of styrene, 5500 g of cyclohexane and 0.75 g of 2,2-bis(2-oxolanyl)propane as a polar substance were charged and the internal temperature of the reactor was raised to



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50°C. A cyclohexane solution containing 0.75 g of n-butyl lithium was fed as a polymerization initiator to the reactor. After the initiation of the reaction, the internal temperature of the reactor gradually increased by the heat generated upon polymerization. Over a period of 5 minutes starting from 7 minutes after the addition of the polymerization initiator and up to 12 minutes after the initiator addition, 105 g of butadiene was fed at a rate of 21 g/min. The final internal temperature in the reactor reached 100°C. Five minutes after the completion of the polymerization reaction, a predetermined amount of tin tetrachloride was added to the reactor and the temperature was kept at 100°C for 10 minutes to carry out a coupling reaction. After addition of an antioxidant to the resulting polymer solution, the solvent was removed, thereby obtaining a styrene-butadiene copolymer (Sample T1) having tin.

As a result of the analysis of Sample T1, the bonding styrene amount was 25 wt.% and the vinyl bond content in butadiene was 39%. It was also found that the weight-average molecular weight (Mw) and the molecular weight distribution (Mw/Mn) in terms of polystyrene as measured by GPC were 450,000 and 1.4, respectively; the coupling ratio determined from the ratio of two peak areas

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of the GPC profile was 48 wt.%; and the modifying ratio determined from GPC using a silica adsorption column was 0%.

Separately, polymerization was carried out in the same manner as for Sample T1. After the polymerization, in place of tin tetrachloride used in the preparation of Sample T1, TGAMH which is a modifier used in the Examples of the present application (including Sample K: see Table 1 of the specification), was added to the obtained polymer in an amount that corresponds to excess epoxy groups with respect to the active ends of the polymer and corresponds to the equivalent amount of epoxy groups with respect to the lithium amount of the fed initiator, to react with each other, thereby obtaining a styrene-butadiene copolymer having a modified component (Sample T2).

As a result of the analysis of Sample T2, the bonding styrene amount was 25 wt.% and the vinyl bond content in butadiene was 39%. It was also found that the weight-average molecular weight (Mw) and the molecular weight distribution (Mw/Mn) in terms of polystyrene as measured by GPC were 455,000 and 1.4, respectively; the coupling ratio determined from the ratio of two peak areas of the GPC profile was 61 wt.%; and the modifying ratio determined from GPC using a silica adsorption column was 55%.

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## 2. Evaluation of Compositions

In the same manner as in Example 3-4 of the present application, rubber compositions were prepared in accordance with the formulation S-1 shown in Table 5 of the specification, except for replacing Sample K used in Example 3-4 with each of Sample T1 and Sample T2 obtained above. The obtained rubber compositions were evaluated in the same manner as the Examples of the present application. The results obtained are shown in the Table below together with the results of Example 3-4.

	Modification ratio	Modifier amount (equivalent/active lithium)	Fuel-cost-saving property ( $\tan\delta$ , 50°C)	Wet skid resistance ( $\tan\delta$ , 0°C)
Sample T1	0	0	0.200	0.505
Sample T2	55	1.54	0.185	0.538
Sample K (Ex. 3-4)	78	0.95	0.138	0.553

From the above results, it can be seen that the rubber composition using Sample T1, which is the same polymer as SBR H of JP '959 and was obtained through a coupling reaction using tin tetrachloride, had extremely deteriorated performances.

In addition, Sample T2 which was obtained using the same base polymer as Sample T1 but modified with an excess amount of the same modifier as used in the Examples

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of the present application had a low modification rate and did not fulfill the claimed modification rate requirement.

As compared to the rubber compositions using Sample T1 and Sample T2, respectively, the rubber composition in Example 3-4 using Sample K show unexpectedly superior effects.

I declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Date: May 15, 2004

Haruo  
Haruo

Yamada  
Yamada